

# Heavy Fermion Scaling: Uranium versus Cerium and Ytterbium Compounds

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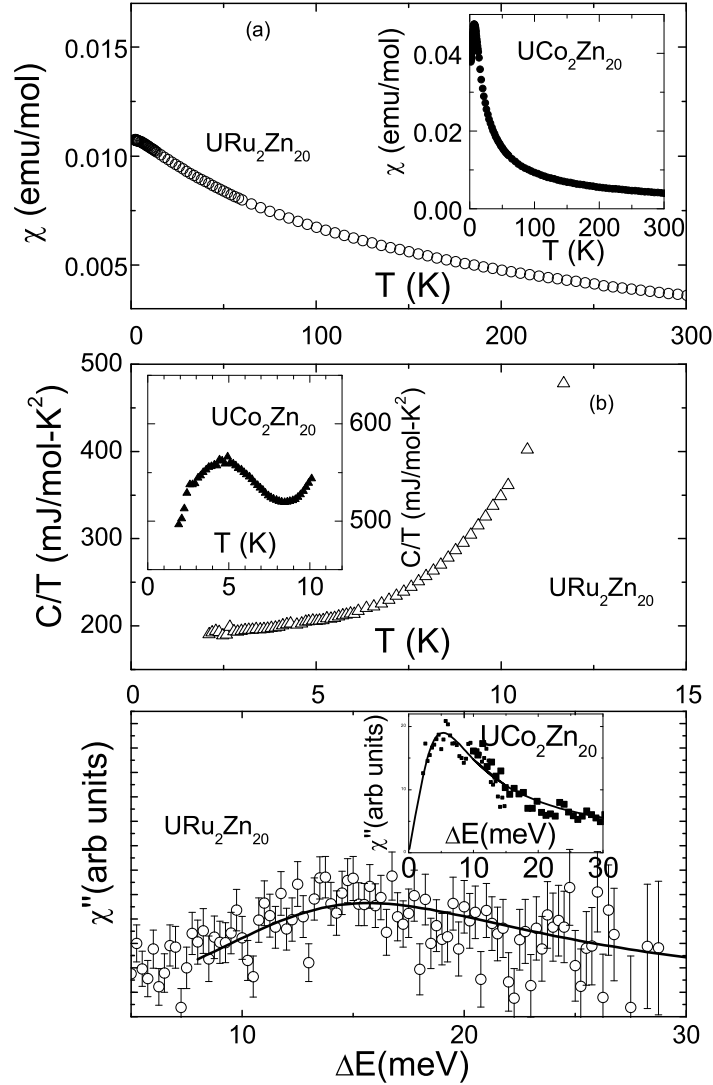
**Abstract.** In an effort to explore the differences between rare-earth-based and uranium-based heavy Fermion (HF) compounds that reflect the underlying difference between local  $4f$  moments and itinerant  $5f$  moments we analyze scaling laws that relate the low temperature neutron spectra of the primary ("Kondo-esque") spin fluctuation to the specific heat and susceptibility. While the scaling appears to work very well for the rare earth intermediate valence compounds, for a number of key uranium compounds the scaling laws fail badly. There are two main reasons for this failure. First, the presence of antiferromagnetic (AF) fluctuations, which contribute significantly to the specific heat, alters the scaling ratios. Second, the scaling laws require knowledge of the high temperature moment degeneracy, which is often undetermined for itinerant  $5f$  electrons. By making plausible corrections for both effects, better scaling ratios are obtained for some uranium compounds. We point out that while both the uranium HF compounds and the rare earth intermediate valence (IV) compounds have spin fluctuation characteristic energies of order 5 - 25 meV, they differ in that the AF fluctuations that are usually seen in the U compounds are never seen in the rare earth IV compounds. This suggests that the  $5f$  itineracy increases the  $f$ - $f$  exchange relative to the rare earth case.

There are two categories of excitation seen in neutron scattering in paramagnetic heavy Fermion compounds. A "primary" spin fluctuation gives rise, when excited, to the high temperature moment. Such an excitation, whose spectrum is typically a broadened Lorentzian, is seen in all such compounds, from moderately heavy intermediate valence (IV) compounds to very heavy Fermion (VHF) compounds. The latter compounds typically reside close to a quantum critical point (QCP) for a  $T = 0$  transition to a magnetic state, where magnetic fluctuations representing critical scattering close to the phase transition also occur.[1] In this situation, scaling laws connect the behavior of the critical fluctuations to that of the specific heat and susceptibility. These scaling laws have a different form for a QCP governed by "local" criticality as compared to the critical behavior of a magnetic instability in a Fermi liquid.[2]

One of the oldest-known properties of VHF and IV materials, whether in uranium or in rare earth (RE) compounds, is the existence of a scaling law whereby the low temperature susceptibility  $\chi(0)$  and specific heat coefficient  $\gamma = C/T$  vary with the inverse  $1/T_{sf}$  of the characteristic energy  $k_B T_{sf}$  of the primary spin fluctuation.[3] The latter quantity can be equated to the maximum  $E_{max}$  in the dynamic susceptibility  $\chi''(E)$ , measured through inelastic neutron scattering. An example is given in Fig. 1, where we compare the susceptibility, specific heat, and neutron spectra of two related compounds,  $\text{URu}_2\text{Zn}_{20}$  and  $\text{UCo}_2\text{Zn}_{20}$ . [4] The zero temperature susceptibility and specific heat coefficient of the latter compound are approximately three times larger, and the energy  $E_{max}$  of the maximum in the neutron spectra is three times smaller than in the former compound.

The scaling laws arising from the primary spin fluctuation receive theoretical justification from the Kondo/Anderson impurity model (K/AIM)[5], where the spin fluctuation temperature  $T_{sf}$  is identified with the Kondo temperature  $T_K$ . Despite the fact that the rare earth atoms form a lattice, the K/AIM works very well to describe the susceptibility  $\chi(T)$ , specific heat  $C(T)/T$ ,  $4f$  occupation number  $n_f$  (related to the valence through  $z = 4 - n_f$  for Ce and  $z = 2 + n_f$  for Yb), as well as the inelastic neutron lineshape for such IV compounds as  $\text{YbAgCu}_4$ [6] and  $\text{YbFe}_2\text{Zn}_{20}$ . [4, 7] A basic condition for this agreement between impurity theory and experiment is that the  $4f$  spin fluctuations, which dominate these measurements, must be nearly localized, or only weakly dependent on momentum transfer  $Q$ . This appears to be the case for such IV compounds as  $\text{YbInCu}_4$ [8],  $\text{YbFe}_2\text{Zn}_{20}$ [4], and  $\text{CePd}_3$ . [9] (Of course the K/AIM is inapplicable to measurements that are highly sensitive to the lattice periodicity, such as electronic transport and de Haas van Alphen measurements.) Because the K/AIM theory works well in this context, we will refer to the primary spin fluctuation as "Kondo-esque."

In the VHF compounds the Kondo-esque fluctuation coexists with magnetic (typically antiferromagnetic or AF) critical fluctuations arising near the QCP, and both contribute to the specific heat and susceptibility. In what follows we will show that although the Kondo-esque scaling initially appears to be invalid in uranium HF compounds, by correcting for the contribution of the AF fluctuations, the scaling



**Figure 1.** (a) Susceptibility, (b) specific heat coefficient and (c) inelastic magnetic neutron scattering spectrum of  $\text{URu}_2\text{Zn}_{20}$ . The insets show the same quantities for  $\text{UCo}_2\text{Zn}_{20}$ . (Data from Ref. [4])

behavior can be restored.

In angle-integrated photoemission studies rare earth  $4f$  states appear as localized states below the Fermi level[10] but uranium  $5f$  states appear as a broad band of emission at the Fermi level.[11] Recent angle-resolved photoemission[12] has determined the dispersion of the  $5f$  states. Essentially, the  $4f$  orbitals are highly localized and hybridize only weakly with the conduction electrons while the  $5f$  orbitals of uranium compounds are spatially extended and form dispersive bands through strong hybridization with the neighboring  $s$ ,  $p$ , and  $d$  orbitals. One of our purposes is to see whether there are differences between the scaling behavior of  $4f$  and  $5f$  heavy Fermion

**Table 1.** Comparison of scaling constants deduced from Kondo theory[13] and the rough phenomenology of the text.

$J$	$E_{max}\gamma(\text{Kondo})$	$E_{max}\gamma(\text{Rough})$	$W(\text{Kondo})$	$W(\text{Rough})$
	$(\pi/3)RJ$	$(3/2) R\ln(2J+1)$	$(1+(1/2J))$	$(2\pi^2/9)/\ln(2J+1)$
3/2	13.0	17.3	1.33	1.58
5/2	21.8	22.3	1.20	1.22
7/2	30.4	25.9	1.14	1.05
9/2	39.1	28.7	1.11	0.95

materials due to the distinction between local and itinerant  $f$  electrons.

The Kondo/Anderson impurity model results in very precise scaling laws, but assumes localized electrons. Since we are concerned with the applicability of scaling to itinerant  $5f$  electrons, we need to stress that the low temperature scaling laws follow from very general considerations. The spin fluctuation peak at  $E_{max} = k_B T_{sf}$  in the dynamic susceptibility represents an excitation of the  $4f$  or  $5f$  moment out of a singlet (non-magnetic) ground state. The excited level has the degeneracy of the local  $4f$  (or itinerant  $5f$ ) moment, which for rare earths is  $N_J = 2J + 1$ . At high temperatures, the moment is excited; at low temperatures, the moment "freezes out" and the system is nonmagnetic.

A phenomenology of the scaling can be obtained by making several simplifying approximations. The spin excitation at  $E_{max}$  will give rise to a peak in the specific heat at a temperature  $T_{max}^C$  that is similar to a Schottky peak, with the exception that  $C(T)$  is linear at low temperature for heavy Fermion compounds. (This linearity is related to the substantial breadth of the excitation.) If we assume that  $C(T)$  is linear for  $T < T_{max}^C$ , that half the  $R\ln(2J+1)$  entropy is generated between  $T = 0$  and  $T_{max}^C$ , and that  $T_{max}^C$  is approximately equal to  $T_{sf}/3$  (a statement that is true for both Schottky anomalies and for the Kondo specific heat), we then obtain  $E_{max}\gamma = 3/2 R\ln(2J+1)$  for the scaling constant. If we further assume that the susceptibility has a van Vleck-like form  $\chi(0) \sim C_J/T_{sf}$  (where  $C_J$  is the free ion Curie constant for total angular momentum  $J$ ) then we obtain the value  $2\pi^2/(9\ln(2J+1))$  for the Wilson ratio  $W = (\pi^2 R/(3C_J)) * \chi(0)/\gamma$ .

It is clear that the scaling constants derived from this phenomenology can only be viewed as roughly approximate. The point of the exercise is that scaling laws with values similar to those of the K/AIM (Table 1) are expected on very general grounds, even when the spin fluctuations arise in an itinerant system where the K/AIM is not expected to be applicable. In particular, the phenomenology exhibits the strong dependence of  $E_{max}\gamma$  on the degeneracy. As we will see, this will allow us to distinguish itinerant from local  $f$  electrons.

In Table 2, we exhibit the experimental values of  $W$  and  $E_{max}\gamma$  deduced from values of  $\chi(0)$ ,  $\gamma$ , and  $E_{max}$  obtained from the literature. In this table, we compare

**Table 2.** Low temperature scaling constants for Ce ( $J = 5/2$ ), Yb ( $J = 7/2$ ), and U ( $J = 4, 9/2$  or undetermined) compounds. Rows marked "Corrected" use the technique described in the text to correct for the AF fluctuation contribution to the specific heat. (Sources of the data given in the first column.)

<i>Compound</i>	$\gamma$ (J/mol-K <sup>2</sup> )	$\chi(0)$ (emu/mol)	$E_{max}$ (K)	$\gamma E_{max}$ (J/mol-K)	Wilson ratio
CePd <sub>3</sub> [20, 21]	0.035	0.0018	638	22.3	1.74
CeSn <sub>3</sub> [22, 23]	0.042	0.0018	464	19.5	1.49
YbAl <sub>3</sub> [24, 25]	0.04	0.005	634	25.4	1.32
YbInCu <sub>4</sub> [6, 8]	0.041	0.006	466	19.1	1.54
YbAgCu <sub>4</sub> [6]	0.199	0.017	133	26.5	0.9
YbFe <sub>2</sub> Zn <sub>20</sub> [4, 7]	0.52	0.05	70	36.4	1.02
UAl <sub>2</sub> [26]	0.14	0.004	243	34.0	0.48
(Corrected)	0.07			17.0	1.30
USn <sub>3</sub> [27, 28]	0.17	0.01	60	10.2	0.99
URu <sub>2</sub> Zn <sub>20</sub> [4]	0.188	0.012	191	35.9	1.08
UPt <sub>3</sub> [29, 14]	0.45	0.009	58	26.1	0.34
(Corrected)	0.225			13.0	1.10
UCo <sub>2</sub> Zn <sub>20</sub> [4, 19]	0.558	0.047	70	39.1	1.42
UBe <sub>13</sub> [29, 30]	1.1	0.015	150	165	0.23
(Corrected)	0.183			27.4	1.38

the uranium HF compounds to the rare earth IV compounds, for the reason that the characteristic temperatures of the spin fluctuations are similar: they are typically 100 K or more, rather than  $\sim 10$ K as seen in the RE VHF compounds. We include all paramagnetic compounds in these two categories for which all three of the quantities  $\chi(0)$ ,  $\gamma$ , and  $E_{max}$  have been reported. It can be seen that the low temperature scaling works well for  $J = 5/2$  cerium and  $J = 7/2$  ytterbium IV compounds. Low temperature scaling also works well for URu<sub>2</sub>Zn<sub>20</sub> and UCo<sub>2</sub>Zn<sub>20</sub>, under the assumption that the high temperature moment is that of a Hund's Rule  $5f^2$  or  $5f^3$  electron. For other classic uranium heavy Fermion compounds such as UAl<sub>2</sub>, USn<sub>3</sub>, UPt<sub>3</sub>, and UBe<sub>13</sub> one or the other of the two scaling constants is unacceptably different (a factor of two or more) from the value expected for the Hund's rule moment.

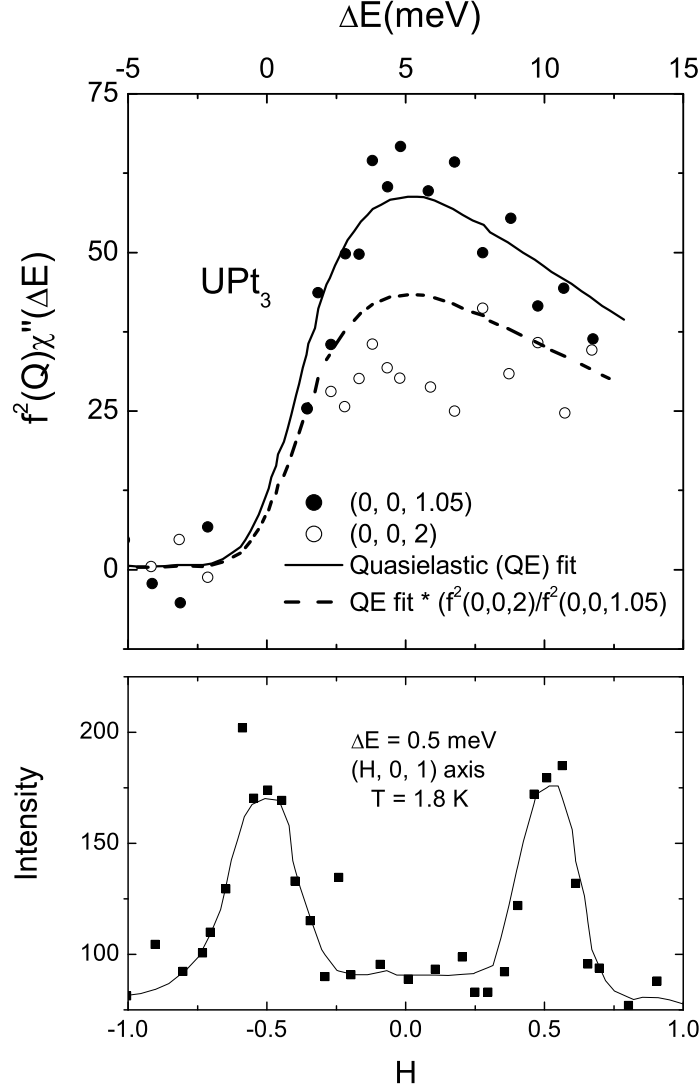
The failure of the scaling laws for these uranium compounds arises from two problems. First, as discussed above, the low temperature specific heat is affected not only by the excitation of the spin fluctuation at  $k_B T_{sf}$  but by antiferromagnetic (AF) fluctuations which occur when the compound resides close to a quantum critical point (QCP) for a  $T = 0$  transition between a magnetic and a nonmagnetic state. Such AF correlations have been observed directly in neutron scattering experiments. These

fluctuations appear as highly  $Q$ -dependent peaks centered near the ordering wavevector  $Q_N$  of the antiferromagnetic state; i.e., they are critical fluctuations. Typically, they occur on an order-of-magnitude lower energy scale than  $k_B T_{sf}$  and are superimposed on a  $Q$ -independent (or weakly  $Q$ -dependent) background of scattering with energy scale  $E_{max} = k_B T_{sf}$ . [14, 15] As an example of this behavior in U compounds, we show data for UPt<sub>3</sub> in Fig. 2. For the scattering on the scale  $E_{max} = 5$ -6 meV, the difference between scattering at zone center (0, 0, 2) and zone boundary (0, 0, 1) can mostly be explained by the uranium  $5f$  form factor. This demonstrates that the primary spin fluctuation has a weak  $Q$ -dependence which, as mentioned above, is known to be the case for RE IV compounds. On the other hand, low energy (0.3 - 0.5 meV) excitations peak sharply near the wavevector (1/2, 0, 1) of the weak antiferromagnetism that occurs in UPt<sub>3</sub>.

These AF fluctuations can give rise to a low temperature upturn in the specific heat coefficient  $C(T)/T$  on the same temperature scale, which enhances the specific heat above the scaling value. The scaling in uranium compounds can be corrected by subtracting this contribution, or rather by extrapolating the specific heat coefficient from temperatures above the upturn. The extrapolation is shown in Fig. 3 for UBe<sub>13</sub>. The low temperature specific heat coefficient  $\gamma = 1.1$  J/mol-K<sup>2</sup> gives  $E_{max}\gamma = 165$  J/mol-K and  $W = 0.23$  for this compound – values which are much too large and small, respectively, compared to the  $J = 4$  or  $9/2$  values of Table 1. If, however, we simply use the extrapolated value  $\gamma = 0.1825$  J/mol-K<sup>2</sup> we obtain  $E_{max}\gamma = 27.4$  J/mol-K and  $W = 1.38$ , values which are in much better accord with the expected scaling constants. Hence, while the extrapolation is not very precise, it does allow us to correct the Kondo-esque scaling values in an obvious manner. [16] Note that while the antiferromagnetic contribution to the specific heat dominates  $\gamma$  at low temperature, the entropy in this contribution (defined as the excess over the extrapolated value) is a small fraction of  $R \ln 2$ . This accords with the fact that the spectral weight of the AF fluctuations seen in the neutron scattering is a small fraction of the weight in the primary spin fluctuation at  $E_{max}$ .

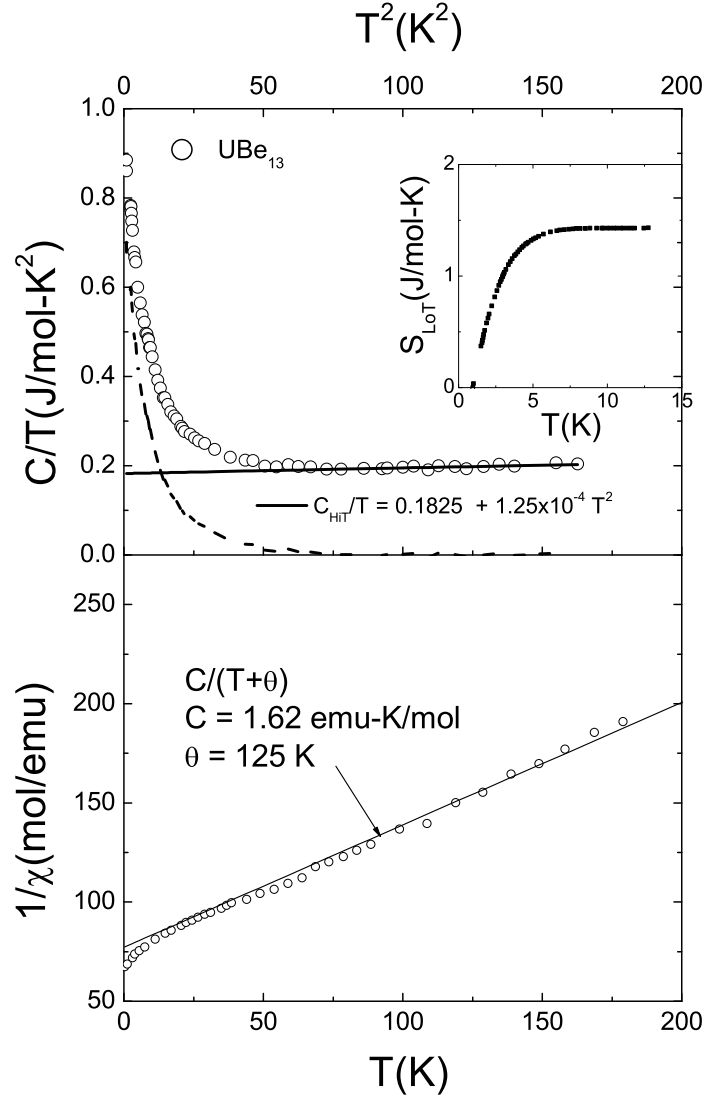
Several other uranium compounds show deviations from the scaling laws. In UPt<sub>3</sub> and UAl<sub>2</sub>, the Wilson ratio is too small compared to the expected value, which should be of order unity, but the scaling constant  $E_{max}\gamma = 26$ -34 is of the right order of magnitude. When the low temperature tail of the specific heat coefficient is subtracted and the extrapolated value of specific heat coefficient (e.g. as in Fig. 4 for UPt<sub>3</sub>) is used in the scaling laws, then the Wilson ratio is corrected, but the  $E_{max}\gamma$  is then too small. We note also that no low temperature tail of the specific heat is observed in USn<sub>3</sub>, and it is also true for that compound that the Wilson ratio is appropriate but  $E_{max}\gamma$  is too small.

We believe that the origin of this discrepancy for these three compounds arises because the degeneracy of the  $5f$  moment, is usually different from the value 9 (or 10) expected for the Hund's rule coupled  $5f^2$  (or  $5f^3$ ) configuration. This can be seen from the fact that the high temperature Curie constant of these compounds (see the inset of Fig. 4a for UPt<sub>3</sub>) is smaller than the free ion value 1.6 emu-K/mol expected for the



**Figure 2.** (a) Intensity versus energy transfer for  $\text{UPt}_3$  at zone center  $(0, 0, 2)$  and close to the zone boundary  $(0, 0, 1.05)$ . The solid line is a fit to a quasielastic power function with  $\Gamma = 5$  meV; the dashed line is the same fit scaled by the ratio of the  $5f$  form factor for the two  $Q$ . (b) Intensity versus Miller index  $H$  along the  $[H, 0, 1]$  direction for energy transfer 0.5 meV. The peaks at  $(1/2, 0, 1)$  represent antiferromagnetic fluctuations. (Adapted from Refs. [14, 15])

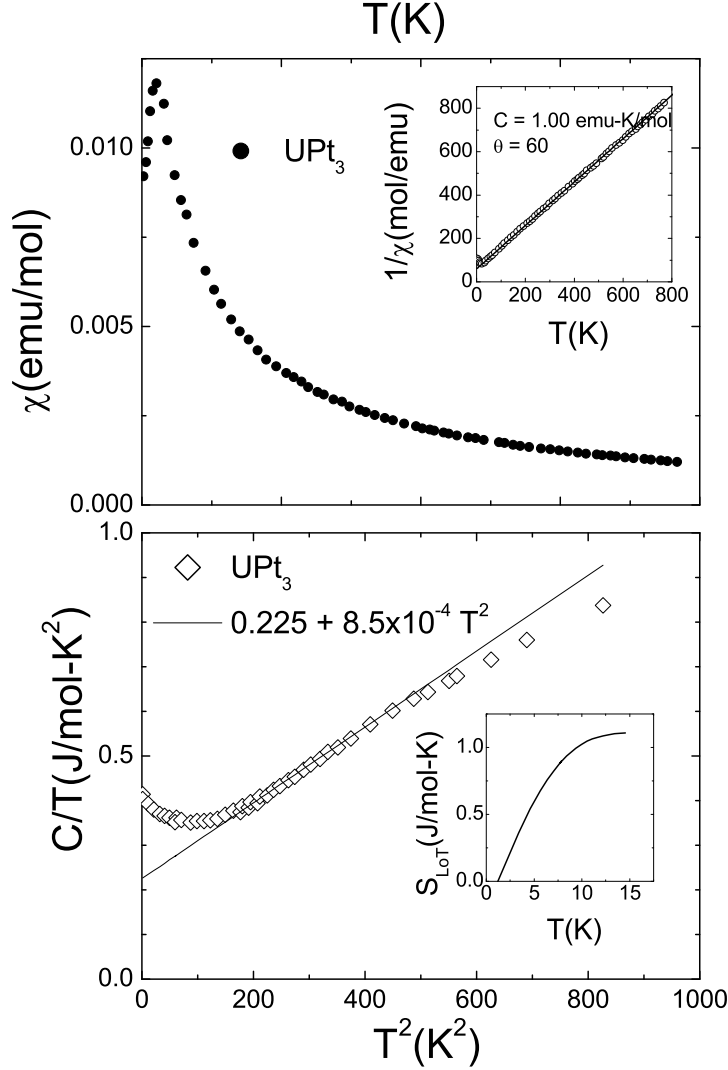
$5f^2$  (or  $5f^3$ ) Hund's rule configurations. To make this statement more graphic, we plot  $T\chi(T)$  in Fig. 5 for a series of uranium compounds. This quantity appears to rapidly approach the Hund's rule limit as temperature is increased for those compounds where the scaling either works well ( $\text{UCo}_2\text{Zn}_{20}$  and  $\text{URu}_2\text{Zn}_{20}$ ) or can be made to work well by correcting for the contribution from the AF fluctuations ( $\text{UBe}_{13}$ ). For the other compounds, it appears to saturate at a smaller value. This suppression of the high temperature moment primarily reflects the suppression of orbital angular momentum,



**Figure 3.** (a) Specific heat coefficient versus the square of the temperature for UBe<sub>13</sub>. The solid line extrapolates from higher temperature; the dashed line is the excess over the extrapolation, due to AF correlations. The entropy in the excess is shown in the inset. (b) The inverse susceptibility; the solid line represents Curie Weiss behavior. (Data from Ref. [29])

which is to be expected for itinerant electrons. Comparison of the  $5f$  form factor between local moment oxides, antimonides, etc. and metallic magnets such as UNi<sub>2</sub> and UFe<sub>2</sub> (Fig. 5 inset) shows that the orbital angular momentum is strongly reduced in the ordered state of the latter compounds.[17] Most heavy Fermion uranium compounds are in a correlated regime intermediate between Hund's rule local moments and purely itinerant uncorrelated  $f$  electron bands; hence we do not know the correct degeneracy to put into the scaling law for  $E_{\text{max}}\gamma$ . (The Wilson ratio is much less sensitive to this degeneracy.) The degeneracy is certainly smaller than  $2J + 1$  for  $J = 4$  or  $9/2$ . This

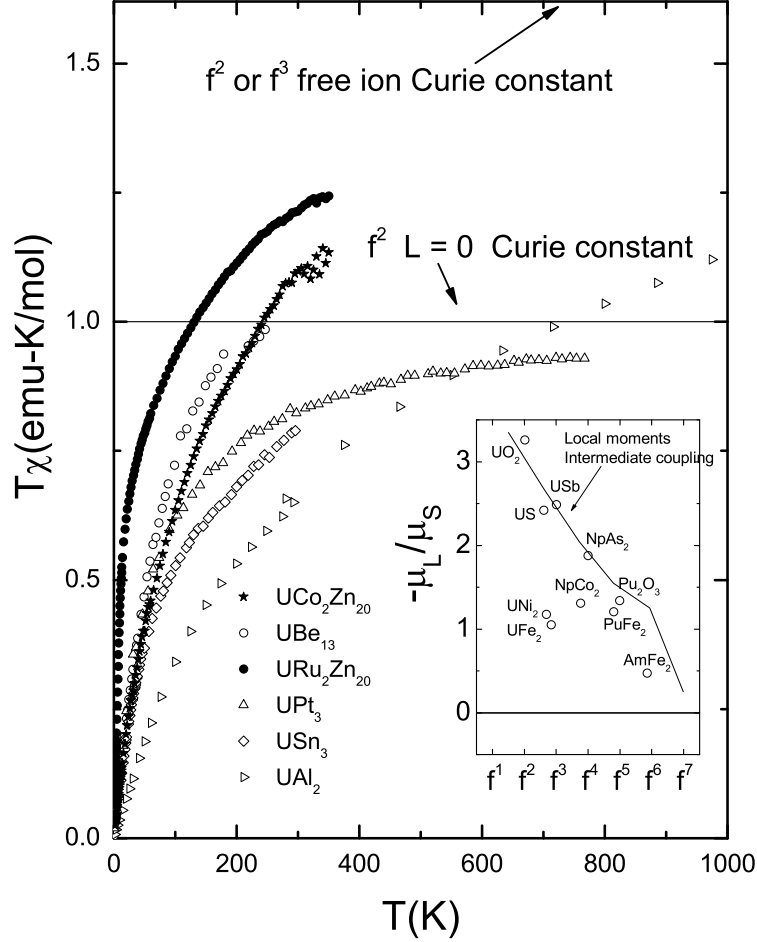




**Figure 4.** (a) Susceptibility of  $\text{UPt}_3$ ; the inverse susceptibility is shown, together with a solid line representing Curie-Weiss behavior in the inset. (b) Specific heat coefficient; the solid line is the extrapolation from high temperature. The entropy in the excess above the extrapolation is shown in the inset. (Data from Ref. [29])

probably explains why the value 13 (17) J/mol-K obtained for this scaling constant for  $\text{UPt}_3$  ( $\text{UAl}_2$ ) after correction for the AF fluctuations (as well as the value 10 seen for  $\text{USn}_3$  without such a correction) is so much smaller than expected for the Hund's rule  $5f^2$  (or  $5f^3$ ) configurations.

This suppression of the degeneracy of the moment associated with the spin fluctuation at  $k_B T_{sf}$  is the primary way that the itineracy of the  $5f$  electrons is reflected in the scaling laws. There are also several differences between the behavior of local moment rare earth-based and itinerant uranium-based heavy Fermion compounds that can be deduced from this analysis. First, we note that the energy scale ( $\sim 100$ -



**Figure 5.** The quantity  $T\chi(T)$  plotted versus temperature for the uranium compounds shown in Table 2. The value of the  $f^2(J = 4)$  and  $f^3(J = 9/2)$  Curie constant is 1.62 emu-K/mol. (Sources for data given in Table 2.) Inset: The ratio of the orbital moment to the spin moment, as determined from measurements of the  $5f$  form factor, for several uranium compounds. Non-metallic compounds yield values of  $\mu_L/\mu_S$  close to the local moment value in intermediate coupling. The intermetallic magnets  $\text{UNi}_2$  and  $\text{UFe}_2$  show strong suppression of the orbital moment. (Data from Ref. [17])

300 K) for the spin fluctuations of the uranium heavy Fermion compounds exhibited in Table 2 is comparable to that of cerium or ytterbium intermediate valence (IV) compounds. However, the AF fluctuations that are seen for most of these uranium compounds are *never* observed in the rare earth IV compounds, but only in very heavy Fermion compounds with small  $T_{sf} \sim 10$  K. It is also true that there is no correlation between the magnitude of the AF contribution and  $T_{sf}$  for the uranium compounds. For

example,  $\text{UBe}_{13}$ , which has a fairly large characteristic energy (150K), shows a large AF contribution to the specific heat, while  $\text{UPt}_3$ , where  $T_{sf}$  is much smaller (60K), exhibits a smaller AF contribution to  $C(T)$  and  $\text{USn}_3$ , for which  $T_{sf} \sim 60\text{K}$ , shows no obvious upturn or AF contribution to  $C(T)$ . [18] A possible explanation of these discrepancies between rare earth and uranium compounds is that the itineracy of the  $5f$  electrons gives rise to an intersite exchange that is larger relative to the spin fluctuation energy in the  $5f$  compounds than in the  $4f$  HF compounds, and which bears no universal relation to  $T_{sf}$ .

From Table 2, it will be noted that the low temperature specific heat, susceptibility, and neutron characteristic energy (and hence the low temperature scaling) are very similar for  $\text{YbFe}_2\text{Zn}_{20}$  and  $\text{UCo}_2\text{Zn}_{20}$ . We have recently [4] compared the data [7, 19] for these two compounds to the predictions of the K/AIM and shown that the theory works extremely well for the rare earth compound, but very poorly for the uranium compound. This disagreement undoubtedly also reflects the difference between local moment and itinerant behavior.

Finally we point to the well-known fact that, unlike the rare earth case, well-defined crystal field excitations are almost never observed in uranium intermetallic compounds. What *is* seen is what we have reported above: a broad excitation on the 5-25 meV scale, representing the primary spin fluctuation, and in some compounds low energy ( $\sim 1$  meV) antiferromagnetic fluctuations. We thus argue that the peak at  $E_{max}$  seen in uranium compounds represents spin fluctuations in an itinerant  $5f$  band.

In conclusion, we have proposed an approach that, while not highly precise, allows for separation of the contribution of the AF fluctuations to the specific heat, thereby giving reasonable values for the Kondo-esque scaling constants. The low values (10-17) of the scaling constant  $E_{max}\gamma$  observed in some uranium HF compounds arise from the low degeneracy of the high temperature moment, which is also reflected in values of the Curie constant which are small compared to the Hund's Rule values. This low degeneracy is a consequence of the itineracy of the  $5f$  electrons.

The results allow us to formulate a line for future research: First, the quantities  $C_p$ ,  $\chi$ , and both  $\chi''_{sf}$  and  $\chi''_{AF}$  (the primary and antiferromagnetic spin fluctuations) should be measured in a single crystal of a given compound. The spectral weight of these two contributions to the magnetic neutron scattering should then be correlated with the fractional entropy of each contribution to the specific heat. Second, the absolute cross section of the primary spin fluctuation should be measured and the appropriate sum rule used to see whether the integrated scattering corresponds to the high temperature moment seen in the susceptibility. Third, the primary spin fluctuation needs to be measured on single crystals of more uranium compounds in an effort to clarify the Q-dependence of this excitation in the coherent state.

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